



(19)

Europäisches Patentamt

European Patent Office

## **Office européen des brevets**



(11) EP 0 524 572 B1

(12)

## EUROPEAN PATENT SPECIFICATION

(45) Date of publication and mention  
of the grant of the patent:  
**02.10.1996 Bulletin 1996/40**

(51) Int. Cl.<sup>6</sup>: **B32B 27/32, B32B 27/36,**  
**B65D 65/40**

(21) Application number: 92112340.2

(22) Date of filing: 20.07.1992

**(54) Stretched, laminated film**

### Gestreckter, laminierter Film

## **Feuille stratifiée et laminée**

**(84) Designated Contracting States:**  
**DE FR GB IT NL**

(30) Priority: 24.07.1991 JP 184860/91

(43) Date of publication of application:  
27.01.1993 Bulletin 1993/04

(73) Proprietor: Mitsubishi Chemical Corporation  
Chiyoda-ku Tokyo (JP)

**(72) Inventors:**

- **Fujii, Toshio**  
**Kurashiki-shi, Okayama-ken (JP)**
  - **Kondo, Toyomitsu**  
**Kurashiki-shi, Okayama-ken (JP)**

- Kato, Kazuhiro  
Kurashiki-shi, Okayama-ken (JP)

(74) Representative: Hansen, Bernd, Dr. Dipl.-Chem.  
et al  
**Hoffmann, Eitle & Partner**  
Patent- und Rechtsanwälte,  
Postfach 81 04 20  
81904 München (DE)

(56) References cited:

**EP-A- 0 204 324**      **FR-A- 2 405 972**  
**US-A- 4 612 221**

- DATABASE WPIL Derwent Publications Ltd., London, GB; & JP-A-4 211 922

**Description****BACKGROUND OF THE INVENTION**

5       The present invention relates to a stretched, laminated film. More particularly, the present invention relates to a stretched, laminated film suitable as a wrapping film which is excellent in cuttability, transparency, heat-resistance, adherence, gas-barrier property, anti-heat-shrinkage, and food safety.

As conventionally known packaging film (wrapping film) used for the purpose of packaging articles, for example, foodstuffs, a film composed of polyvinylidene chloride, polyethylene or polyvinyl chloride as the main constituent can be 10 exemplified. These wrapping films in the state of packaging the articles are often charged into an electronic oven so as to be heated, so that they are required not only to have good adherence but also to be prevented from heat-shrinkage, melting or color change when heated to a high temperature, in other words, to have an excellent heat resistance.

However, a polyvinylidene chloride film is disadvantageous in that the heat-shrinkage is large. A polyethylene film has a defect in that the film melts when contacts with a hot oily matter such as hot meat and fried food. A polyvinyl chloride film suffers from a problem of a whitening phenomenon which occurs when contacts with boiling water.

As resins for a packaging film having a heat-resistance, polyesters such as a polyethylene terephthalate (hereinafter referred to as "PET") are known. PET has both an excellent heat-resistance and an excellent mechanical strength, but it is difficult to impart adherence to a PET. It is, therefore, difficult to apply PET to a wrapping film in the form of a single-layer film. A wrapping film is required to have an excellent cuttability. A wrapping film is generally contained in a case such as a paper box and the film is drawn out of the case by a necessary length and cut off with the target end of the film brought into contact with a cutting blade so-called "a cutting edge" which is attached to the case. As the "cutting edge" used for cutting off the film, a simple blade produced merely by blanking an iron plate of about 0.2 mm in thickness into the shape of a saw is generally used. The case holding the "cutting edge" is generally a paper box made of coated board having a strength of about 350 to 700 g/m<sup>2</sup>, so that the rigidity thereof is very low.

25      This type of film is required to be easily cut by such a simple cutting mechanism as desired by the user. However actually, not only the case or the "cutting edge" is sometimes deformed but also the film is sometimes deformed or cut at a position deviated from the "cutting edge". Concretely, at the time of cutting, when a rift occurs on a conventional polyvinylidene film, the film has a tendency of being cut obliquely along the rift instead of along the "cutting edge". A conventional polyethylene film requires a considerably much pulling force at the time of cutting, so that the case is sometimes bent and the film is sometimes deformed.

Furthermore, a wrapping film which does not generate chlorine or the like during use or when burned, has recently been demanded in consideration of environmental problems.

A packaging film comprising an inner layer and an outer layer composed of polypropylenes composition, and an intermediate layer composed of aliphatic polyamides was proposed as a wrapping film which generates no chlorine 35 (Japanese Patent Application Laid-Open (KOKAI) Nos. 2-321605, 3-13550, 3-15547, and 3-283346).

In spite of the proposal of these films, a laminated film composed of a resin containing no chlorine atom in a polymer molecule, and which has more excellent heat-resistance, adherence, gas-barrier property and easy cutting property is strongly demanded.

40      As a result of various studies undertaken by the present inventors so as to solve this problem, it has been found that by stretching a laminate comprising polyesters such as a PET resin as an intermediate layer and a polypropylenes composition as an outer layer and an inner layer composed of 100 parts by weight of polypropylenes, 2 to 30 parts by weight of polybutene and/or polyisobutylene and 0.2 to 5 parts by weight of a polyglycerin fatty acid ester, the resultant stretched, laminated film has excellent cuttability, transparency, heat-resistance, adherence, gas-barrier property, anti-heat shrinkability and food safety, especially excellent heat-resistance and cuttability without impairing the intrinsic properties (heat-resistance and mechanical strength) of a polyester resin, and further is favorable in the view-point of the environmental problems. The present invention has been achieved on the basis of this finding.

**SUMMARY OF THE INVENTION**

50      In an aspect of the present invention, there is provided a stretched, laminated film comprising at least an outer layer, an intermediate layer and an inner layer,

the outer layer and inner layer comprising a polypropylenes composition composed of 100 parts by weight of polypropylenes (A), 2 to 30 parts by weight of polybutene and/or polyisobutylene (B) and 0.2 to 5 parts by weight of a polyglycerin fatty acid ester (C), and

55      the intermediate layer comprising polyesters (D) containing ethylene terephthalate units as the main component; and

having a shear peel strength of the outer layer and the inner layer of not less than 0.4 kg/cm<sup>2</sup>, and a 180° peel strength thereof of not more than 4 g/50 mm,

a tensile break elongation in the machine direction (= direction parallel to the direction of drawing) of not more than 200%.

a ratio of the tensile break elongation in the machine direction (= direction parallel to the direction of drawing) and that in the transverse direction (= direction vertical to the direction of drawing) of not more than 3,

5 a thickness of not more than 40 µm, and

a thickness of the intermediate layer of 10 to 90% of the total film thickness.

#### DESCRIPTION OF THE PREFERRED EMBODIMENTS

10 In the polypropylenes composition in the present invention, a larger amount of polypropylenes (A) than either of the polybutene and/or polyisobutylene (B) component and the polyglycerin fatty acid ester (C) component is contained, and the polypropylenes (A) is used as the base material of the composition.

As the polypropylenes (A), for example, a propylene homopolymer and a copolymer of propylene and a small amount of another comonomer such as ethylene and butene are usable. Examples of the propylene homopolymer are 15 an isotactic polypropylene, a syndiotactic polypropylene and an atactic polypropylene. Among these, an isotactic polypropylene is preferable. As the copolymer of propylene and another comonomer such as ethylene and butene (content: 0.5 to 7 wt%), a random copolymer and a block copolymer are both usable, but a propylene-ethylene random copolymer containing 1 to 5 wt% of ethylene is preferable. The physical properties of the polypropylenes (A) may be appropriately selected in accordance with the purposes of use and conditions, but the polypropylenes (A) having a melt flow rate 20 (MFR) of 0.5 to 20 g/10 min (measured at a temperature of 230°C and with a load of 2.16 kg in accordance with JIS K 6758), more preferably 3 to 15 g/10 min, a density of 0.89 to 0.91 g/cm<sup>3</sup>, more preferably 0.893 to 0.905 g/cm<sup>3</sup> are preferable.

The polybutene and/or polyisobutylene (B) in the polypropylenes composition acts as an adhesion component imparting an excellent adherence to the film produced. The physical properties of the polybutene and/or polyisobutylene (B) is not specified and may be appropriately selected in accordance with the amount, the kind of another copolymer component, and the purposes of use of the target composition. It is generally preferable that the polybutene and/or polyisobutylene (B) has a number-average molecular weight of 200 to 3,000, more preferably 400 to 2,000, and a viscosity of 25 300 to 50,000 cps, more preferably 500 to 15,000. The amount of polybutene and/or polyisobutylene (B) is 2 to 30 parts by weight, preferably 5 to 25 parts by weight based on 100 parts by weight of the polypropylenes (A). If the amount of polybutene and/or polyisobutylene (B) is less than 2 parts by weight, it is difficult to impart a sufficient adherence to the film. On the other hand, if the amount exceeds 30 parts by weight, stickiness becomes to be disadvantageously caused by a large amount of bleeding, and the heat-resistance becomes to be lowered.

The polyglycerin fatty acid ester (C) in the polypropylenes composition used together with the polybutene and/or polyisobutylene (B) increases the adherence of the film. The polyglycerin fatty acid ester (C) is a compound obtained 35 by esterifying at least one hydroxyl group of a glycerin condensation with a higher fatty acid having 8 to 22 carbon atoms. The polymerization degree of the glycerin condensation is generally 2 to 10, preferably 2 to 6, more preferably 2 to 3. The number of hydroxyl groups of the glycerin condensation which are esterified is at least one, preferably at least one and not more than 70% based on the total number of hydroxyl groups, more preferably at least one and not more than 60% based on the total number of hydroxyl groups. The higher fatty acid may be either saturated or unsaturated fatty acid having 8 to 22 carbon atoms. A higher fatty acid having 10 to 18 carbon atoms is particularly preferable. As examples of the polyglycerin fatty acid ester (C), diglycerin monolaurate, diglycerin monopalmitate, diglycerin monooleate, diglycerin monostearate, diglycerin dioleate and triglycerin monooleate may be exemplified. Among these, diglycerin monooleate and diglycerin monolaurate are preferable.

Two or more of these polyglycerin fatty acid ester (C) may be jointly used. The amount of polyglycerin fatty acid 45 ester (C) used is 0.2 to 5 parts by weight, preferably 0.5 to 3 parts by weight based on 100 parts by weight of the polypropylenes (A). If the amount of polyglycerin fatty acid ester (C) is less than 0.2 part by weight, it is difficult to produce a synergistic effect of the polypropylenes (A) and the polyglycerin fatty acid ester (C). On the other hand, if the amount exceeds 5 parts by weight, stickiness and odor become to be disadvantageously caused by a large amount of bleeding and a large amount of gel unfavorably tends to be produced.

50 The polyester (D) constituting the intermediate layer of the wrapping film of the present invention is a thermoplastic polyester obtained by the polycondensation of a dibasic acid and a divalent alcohol. For example, a saturated polyester produced from terephthalic acid or a derivative thereof and ethylene glycol is preferable.

As the terephthalic acid derivative as the main acid component of the polyester (D), a lower alkyl (C<sub>1</sub>-C<sub>8</sub>) ester of 55 terephthalic acid is preferable. An alkyl ester having 1 to 4 carbon atoms, particularly, dimethyl terephthalate is preferable.

In the present invention, a part (generally 2 to 20 wt%) of the acid component may be replaced by another acid component such as dicarboxylic acid or oxycarboxylic acid other than terephthalic acid, for example, isophthalic acid, phthalic acid, 2,6-naphthalenedicarboxylic acid, parahydroxyethoxybenzoic acid, adipic acid, sebacic acid, and a lower alkyl (C<sub>1</sub>-C<sub>8</sub>) ester thereof.

The main glycol component of the polyester (D) in the present invention is ethylene glycol. A part (generally 2 to 20 wt%) of the ethylene glycol may be replaced by another glycol component such as trimethylene glycol, tetramethylene glycol, hexamethylene glycol, neopentyl glycol and 1,4-cyclohexanedimethanol.

In any case, the polyester (D) in the present invention refers to a polyester containing ethylene terephthalate units as the main component, for example, not less than 80 mol% based on all the components.

As the polyester (D) in the present invention, polyethylene terephthalate (PET) composed of about 50 mol% of terephthalic acid and about 50 mol% of ethylene glycol, and having a viscosity-average molecular weight of 10,000 to 30,000, preferably 17,000 to 22,000 is particularly preferable because it has excellent transparency and film-forming property, and good heat-resistance which are required of a wrapping film.

10 The appropriate intrinsic viscosity  $[\eta]$  of the polyester (D) of the present invention is 0.4 to 1.0, preferably 0.5 to 0.8 from the view-point of the film-forming property.

The wrapping film of the present invention is produced by forming an unstretched, laminated film composed of the polypropylenes composition as the outer and inner layers and the polyester (D) as the intermediate layer by any conventional lamination method, and stretching the resultant unstretched, laminated film.

15 As examples of a method of producing the unstretched, laminated film (i.e. lamination method), a heat-bonding lamination method, an extrusion lamination method, a dry lamination method and a coextrusion method may be exemplified. Among these, a coextrusion method is particularly preferable.

As the coextrusion method, a T-die extrusion method and an air-cooling or water-cooling inflation method may be adopted. Among these, a T-die extrusion method and a water-cooling inflation method are preferable. By quenching the 20 melt-extruded resin by either of these methods, it is possible to obtain a film having an excellent transparency.

The unstretched, laminated film is formed so that the outer layer and the inner layer have a thickness of 10 to 200  $\mu\text{m}$ , preferably 20 to 100  $\mu\text{m}$ , and the intermediate layer has a thickness of 5 to 200  $\mu\text{m}$ , preferably 10 to 50  $\mu\text{m}$ . An adhesive layer may be disposed between the intermediate layer and the inner layer and/or between the intermediate layer and the outer layer, and the thickness of the adhesive layer is 2 to 40  $\mu\text{m}$ , preferably 4 to 20  $\mu\text{m}$ .

25 The unstretched, laminated film obtained by a coextrusion method is stretched in at least one direction, namely, either in the machine direction (parallel to the direction of film drawing) or in the transverse direction (vertical to the direction of film drawing). The film is uniaxially or biaxially stretched by a stretch ratio of 2 to 6 times, preferably 3 to 5 times in each direction. In the case of uniaxial stretching, the film is preferably stretched in the machine direction by the above described stretch ratio. If the stretch ratio is less than 2, the cuttability of the stretched film tends to be lowered.

30 If the stretch ratio is more than 6, the stretching properties of the film tend to be lowered and a rupture and an unevenness of stretching are apt to be generated. The stretching operation is carried out, for example, by heating the unstretched film as it is or after slit to a predetermined width, stretching the heated film in the machine direction by, for example, changing the peripheral speed of stretch rolls, and then stretching the resultant film in the transverse direction by a tentering stretching method. The preheating temperature for the stretching process is in the range of 50°C to (melting point of the polypropylene (A) - 10°C), preferably in the range of 70 to 140°C.

35 The heat-setting temperature after stretching the film is a temperature higher than the preheating temperature. In order to prevent the film from wrinkling and shrinking at a high temperature, the film is preferably heat-set at as high a temperature as possible. The heat-setting process is carried out at a temperature in the range of the preheating temperature to (melting point of the polypropylene (A) - 10°C), preferably in the range of 80 to 150°C.

40 If the preheating temperature or the heat-setting temperature is over a temperature of about 10°C lower than the melting point of the polypropylene (A), the film easily adheres to the stretch rolls. On the other hand, if the preheating temperature is lower than 50°C, the PET tends to become difficult to stretch due to the insufficient heat, thereby tending to cause a rupture.

The thickness of the packaging film of the present invention is not more than 40  $\mu\text{m}$ , preferably 2 to 30  $\mu\text{m}$ , more 45 preferably 5 to 20  $\mu\text{m}$ . The thickness of the intermediate layer is 10 to 90%, preferably 15 to 80%, more preferably 20 to 40% based on the total thickness. If the film thickness is larger than 40  $\mu\text{m}$ , the tear strength of the film, when the film is cut, becomes too large for a wrapping film.

If the thickness of the intermediate layer is less than 10% based on the total thickness, it is difficult to maintain the 50 intrinsic heat-resistance and mechanical strength of PET. The intermediate film, preferably, has a thickness of not less than 15% based on the total thickness. On the other hand, if the thickness of the intermediate film is larger than 90% based on the total thickness, the layer of the polypropylene layer becomes so thin that it is difficult to film-form the intermediate layer uniformly by coextrusion. In addition, it is disadvantageous from the view-point of cost. The intermediate layer preferably has a thickness of not more than 80% based on the total thickness.

The stretched, laminated film of the present invention is composed of at least three resin layers, namely, outer 55 layer/intermediate layer/inner layer. The stretched, laminated film of the present invention may have four or more resin layers. For example, the film may have a structure of outer layer/adhesive layer/intermediate layer/inner layer, outer layer/intermediate layer/adhesive layer/inner layer, or outer layer/adhesive layer/intermediate layer/adhesive layer/inner layer.

In the present invention, a five-layered film composed of three kinds of resins having an adhesive layer of a modified polyolefin between the outer layer and the intermediate layer, and between the inner layer and the intermediate layer is especially preferable, because it does not impair the transparency and the film-forming property as a wrapping film. As examples of the modified polyolefin, a polyolefin such as polyethylene and polypropylene having a polar group of an unsaturated carboxylic acid such as anhydrous maleic acid or a derivative thereof added thereto by a graft reaction may be used. As a resin to be grafted, the polyester (D) used for the inner and outer layers is preferable in the point of adherence.

The amount of modifying monomer of an unsaturated carboxylic acid, particularly, anhydrous maleic acid to be grafted is 0.01 to 10 parts by weight, preferably 0.01 to 3 parts by weight based on the parts by weight of the base resin to be grafted from the view-point of an adhesion with the polyester (D) as the intermediate layer.

It is preferable to dispose such an adhesive layer so as to have a thickness of 2 to 20%, preferably 3 to 10% based on the total thickness of the wrapping film.

Alternatively, by adding an appropriate adhesive component to each layer, it is possible to prevent each layer of a three-layer structure from peeling off without disposing any adhesive layer.

As examples of the adhesive component to be added to the outer layer, the intermediate layer and the inner layer (the amount of adhesive component to be added: 5 to 20 wt%), Novatec AP790P, AP792P, AP196P and AP197P (mfd. by Mitsubishi Kasei Corporation) may be exemplified.

The packaging film produced in the above-described manner has a shear peel strength of not less than 0.4 kg/cm<sup>2</sup>, preferably 0.5 to 2.5 kg/cm<sup>2</sup>, and a 180° peel strength of not more than 4 g/50 mm, preferably 1 to 3 g/50 mm. The tensile break elongation in the machine direction (parallel to the direction of drawing) is not more than 200%, preferably 5 to 150%, and the ratio of the tensile break elongation in the machine direction (parallel to the direction of drawing) and that in the transverse direction (vertical to the direction of drawing) is not more than 3, preferably 0.1 to 2.

The heat-resistant temperature of the wrapping film of the present invention is not lower than 200°C, preferably not lower than 210°C, and the percentage of heat-shrinkage at a temperature of 140°C for 45 sec is not more than ±5%, preferably not more than ±3.5% both in the machine direction and in the transverse direction. The gas barrier property of the wrapping film of the present invention is not more than 500 cc/m<sup>2</sup> • 24 hr • atm.

These physical properties are obtained by the later-described measuring methods, and the shear peel strength and the 180° peel strength represent the adherence of the wrapping film of the present invention, and the tensile break elongation and the ratio of the tensile break elongation represent the cuttability.

The wrapping film of the present invention has excellent cuttability, transparency, heat-resistance, adherence, gas-barrier property, anti-heat shrinkage and food stability.

In addition, the wrapping film of the present invention is free from the problem of generation of chlorine, so that it is also favorable from the view-point of the environmental problems.

### 35 Examples

The present invention will be explained in more detail hereinunder with reference to the following examples.

The physical properties of the film obtained were measured by the following methods.

#### 40 (1) Transparency (haze value)

The transparency of the film was measured by a haze meter in accordance with JIS K 6714. The haze value of the stretched, laminated film of the present invention of not more than 5, more preferably not more than 3 is preferred.

#### 45 (2) Heat-resistant temperature

Paper of 25 mm in length was applied to both the upper and lower ends of a test piece cut into a strip of 30 mm in width and 14 cm in length, and a weight of 10 g was hung from the test piece for 1 hour. The maximum ambient temperature at which the test piece was not broken, was read in the unit of ten degrees (See "Quality indication of wrapping film", the Tokyo-regulation No. 1072).

#### (3) Percentage of heat-shrinkage

The film was cut into a test piece 10 mm in diameter by a blanking jig, and the test piece was dipped in an aluminum pan accommodating silicone oil. The aluminum pan was placed on a hot plate heated to 140°C. The temperature of the hot plate was temporarily lowered because the heat was transferred to the aluminum pan. The test piece is maintained at a temperature of 140°C for 45 sec after the temperature of the hot plate rose again to 140°C, and then the test piece was taken out and a change in the size thereof was measured. The result is expressed in percentage.

(4) Cuttability

5 The film was wound around a tubular core and accommodated in a wrapping film case. The film was cut with the cutting edge, and the cuttability was evaluated in four levels (good, normal, slightly bad, bad) on the assumption that the cuttability of a commercially available wrapping film was in the level of "normal". The wrapping film of the present invention is required to have a cuttability of "good".

(5) Adhesion strength

10 Two sheets of film were laid with one on top of the other. A roll of 25 mmf and 150 mm in width was pressed against the film at a linear pressure of 0.3 kg/cm three times. Within 5 minutes thereafter, the forces required for shear peeling the two sheets of film and for peeling the two sheets of film in the direction of 180° were measured by an Instron-type tensile strength tester under the conditions shown in Table 1.

15

Table 1

	Shear peeling	180° peeling	
20	Pulling speed	300 mm/min	300 mm/min
	Chuck gap	50 mm	50 mm
25	Width of sample	25 mm	50 mm
	Area for adhesion	25 mm × 25 mm	50 mm × 50 mm
	Unit of measured value	kg/cm <sup>2</sup>	g/50 mm

(6) Tensile break elongation and the ratio of the tensile break elongation

30 The tensile break elongations of the film in the machine direction (parallel to the direction of drawing) and the transverse direction (vertical to the direction of drawing) were measured in accordance with JIS C 2318. The ratio of the tensile break elongations in the machine direction and in the transverse direction was obtained from these values.

35

Examples 1 to 4, Comparative Examples 1 to 8

40 A propylene-ethylene random copolymer [Mitsubishi Polypro 6500J (trade mark), produced by Mitsubishi Kasei Corporation] (MFR: 9.0, density: 0.896, ethylene content: 2.5%) was used as the polypropylene (A), polybutene [Nissan Polybutene 015SH (trade name), produced by Nippon Oil and Fats Co., Ltd.] (average molecular weight: 580, viscosity: 2,000 cps at 25°C) was used as the polybutene and/or polyisobutylene (B), and diglycerin monooleate (produced by Riken Vitamin Co., Ltd.) was used as the polyglycerin fatty acid ester (C). The amount of each component used is shown in Table 2, and these components were mixed by a blender and thereafter the resultant mixture was pelletized by a biaxial extruder having a diameter of 30 mm at a temperature of 220°C to produce a polypropylene composition.

45

45 By using a three-kind five-layer T-die coextruder having a width of 450 mm, the polypropylene composition was charged into an extruder No. 1 having a diameter of 50 mm, a polyethylene terephthalate having a viscosity-average molecular weight of 20,000 and an intrinsic viscosity  $[\eta]$  of 0.60 dl/g (produced by Mitsubishi Kasei Corporation) was charged into an extruder No. 2 having a diameter of 40 mm as the polyester (D), and an adhesive polyolefin resin, Novatec-AP196T (trade name, produced by Mitsubishi Kasei Corporation) was charged into an extruder No. 3 having a diameter of 40 mm. These components were simultaneously extruded at a die temperature of 280°C and formed into a film at a chill roll temperature of 30°C and a draw-off speed of 20 m/min to produce a three-kind five-layered film of 170 µm in thickness which is composed of: outer layer (polypropylene composition)/adhesive layer (adhesive resin)/intermediate layer (PET)/adhesive layer (adhesive resin)/inner layer (polypropylene composition). The thickness ratio of the layers of each film is shown in Table 2.

55

55 The film obtained was stretched in the machine direction (MD) by rolls and in the transverse direction (TD) by a tenter by a stretch ratio shown in Table 2 at a preheating temperature of 90°C and was heat-set at a heat-setting temperature of 120°C so as to produce a stretched multi-layer film of 10 µm in thickness.

The transparency, heat-resistant temperature, percentage of shrinkage, cuttability and adhesion strength were measured. The results are shown in Table 3.

EP 0 524 572 B1

The physical properties of the films in Comparative Examples 2 and 5 were unable to be evaluated respectively, because stripes were produced due to the addition of excess polyglycerin fatty acid ester (C) in Comparative Example 2, and a because rupture was caused during the stretching process, which made sampling impossible, in Comparative Example 5.

5 The cuttability of the films in Comparative Examples 6 and 8 were evaluated to be "slightly bad" and "bad", respectively, because the films extended slightly and were difficult to cut.

Table 2

		Composition of PP layer (parts by weight)			Thickness ratio of layers (PP/Ad/PET/ Ad/PP)	Stretch ratio (MD/TD)
		(A)	(B)	(C)		
Example	1	100	12	1	3/1/2/1/3	4/4
	2	100	12	2	3/1/2/1/3	4/4
	3	100	14	1	3/1/2/1/3	4/4
	4	100	12	2	3/1/2/1/3	3.5/1
Comparative Example	1	100	12	0	3/1/2/1/3	4/4
	2	100	12	6	3/1/2/1/3	1/1
	3	100	2	2	3/1/2/1/3	4/4
	4	100	35	2	3/1/2/1/3	4/4
	5	100	12	2	3/1/2/1/3	7/7
	6	100	12	2	3/1/2/1/3	1.7/1.7
	7	100	12	2	3/1/2/1/5	4/4
	8	100	12	2	3/1/2/1/3	1.7/4

35

40

45

50

55

Table 3

	Haze (%)	Heat-resistant temp. (°C)	Percentage of heat-shrinkage MD/TD (%)	Cutability	Adhesion strength 180° peeling (kg/cm <sup>2</sup> ) (g/50mm)	Tensile break elongation MD (%)	Tensile break elongation TD (%)	Ratio (MD/TD)		
Example 1	0.6	230	2/0	Good	0.52	1.5	125	77	1.6	
	2	0.3	230	2/0	Good	0.56	1.7	128	80	1.6
	3	0.6	230	2/0	Good	0.93	2.1	125	80	1.6
	4	0.6	230	3/-1	Good	0.50	1.7	71	410	0.2
Comparative Example 1	0.7	230	2/0	Good	0.33	1.2	126	78	1.6	
	2	-	-	-	-	-	-	-	-	
	3	0.8	230	2/0	Good	0.15	0.91	130	80	1.6
	4	0.4	230	2/0	Good	2.9	5.0	127	81	1.6
	5	-	-	-	-	-	-	-	-	
	6	2.0	230	2/0	Slightly bad	0.54	1.8	161	108	1.5
	7	0.7	170	23/19	Good	0.60	1.8	152	53	2.9
	8	0.4	230	0/1	Bad	0.55	1.5	153	72	2.1

**Claims**

1. A stretched, laminated film comprising at least an outer layer, an intermediate layer and an inner layer,  
 5        said outer layer and inner layer comprising a polypropylenes composition composed of 100 parts by weight of polypropylenes (A), 2 to 30 parts by weight of polybutene and/or polyisobutylene (B) and 0.2 to 5 parts by weight of a polyglycerin fatty acid ester (C); and  
 10      said intermediate layer comprising polyesters (D) containing ethylene terephthalate units as the main component;  
 15      said film being uniaxially or biaxially stretched by a stretch ratio of 2 to 6 times in each direction; and  
 said film having a shear peel strength of said outer layer and said inner layer of not less than 0.4 kg/cm<sup>2</sup>, and a 180° peel strength thereof of not more than 4 g/50 mm, (measured as stated in the description),  
 a tensile break elongation in the machine direction of not more than 200% (measured according to JIS C 2318),  
 15      a ratio of the tensile break elongation in the machine direction and that in the transverse direction of not more than 3,  
 a thickness of not more than 40 µm, and  
 a thickness of said intermediate layer of 10 to 90% of the total film thickness.
- 20     2. A stretched, laminated film according to claim 1, wherein said polypropylenes (A) are either of a propylene homopolymer and a propylene copolymer.
3. A stretched, laminated film according to claim 1, wherein said polypropylenes (A) have a melt flow rate of 0.5 to 20 g/10 min and a density of 0.89 to 0.91 g/cm<sup>3</sup>.
- 25     4. A stretched, laminated film according to claim 1, wherein said polybutene and/or polyisobutylene (B) has a number-average molecular weight of 200 to 3,000 and a viscosity of 300 to 50,000 cps.
- 30     5. A stretched, laminated film according to claim 1, wherein said polyglycerin fatty acid ester (C) is a compound obtained by esterifying at least one hydroxyl group of a glycerin condensation with a higher fatty acid having 8 to 22 carbon atoms.
- 35     6. A stretched, laminated film according to claim 1, wherein said component (D) is a saturated polyester produced from terephthalic acid or a derivative thereof and ethylene glycol.
7. A stretched, laminated film according to claim 1, further comprising an adhesive layer between said intermediate layer and said outer layer and between said intermediate layer and said inner layer.
- 40     8. A stretched, laminated film according to claim 7, wherein said adhesive layer comprises modified polyolefins.
9. A stretched, laminated film according to claim 1, wherein said film is a coextruded, laminated film.
- 45     10. A stretched, laminated film according to claim 1, wherein said film is obtained by biaxially stretching an unstretched laminated film in the machine direction and in the transverse direction.
11. A stretched, laminated film according to claim 1, wherein said film is obtained by uniaxially stretching an unstretched laminated film in the machine direction.

**Patentansprüche**

- 50     1. Gestreckter Schichtfilm, der wenigstens eine äußere Schicht, eine Zwischenschicht und eine innere Schicht umfaßt,  
 wobei besagte äußere Schicht und innere Schicht eine Polypropylenzusammensetzung umfassen, die aus 100 Gewichtsteilen Polypropylen (A), 2 bis 30 Gewichtsteilen Polybuten und/oder Polyisobutylein (B) und 0,2 bis 5 Gewichtsteilen eines Polyglycerin-Fettsäureesters (C) zusammengesetzt ist; und  
 besagte Zwischenschicht Polyester (D) umfaßt, die Ethylenterephthalateinheiten als die Hauptkomponente enthalten; und

besagter Film einaxial oder biaxial gestreckt ist mit einem Streckverhältnis von 2 bis 6 mal in jeder Richtung; und

5 besagter Film eine Scher-Schälfestigkeit besagter äußerer Schicht und besagter innerer Schicht von nicht weniger als 0,4 kg/cm<sup>2</sup> und eine 180° Schälfestigkeit derselben von nicht mehr als 4g/50 mm (gemessen wie in der Beschreibung angegeben),

10 eine Zug-Bruchdehnung in der Maschinenrichtung von nicht mehr als 200 % (gemessen in Übereinstimmung mit JIS C 2318),

15 ein Verhältnis der Zug-Bruchdehnung in der Maschinenrichtung und der in der Querrichtung von nicht mehr als 3,

15 eine Dicke von nicht mehr als 40 µm, und

15 eine Dicke besagter Zwischenschicht von 10 bis 90 % der gesamten Filmdicke hat.

2. Gestreckter Schichtfilm nach Anspruch 1, dadurch gekennzeichnet, daß besagte Propylene (A) entweder ein Propylenhomopolymer oder ein Propylencopolymer sind.

20 3. Gestreckter Schichtfilm nach Anspruch 1, dadurch gekennzeichnet, daß besagte Polypropylene (A) eine Schmelzflußrate von 0,5 bis 20 g/10 Min. und eine Dichte von 0,89 bis 0,91 g/cm<sup>3</sup> haben.

25 4. Gestreckter Schichtfilm nach Anspruch 1, dadurch gekennzeichnet, daß gesagtes Polybuten und/oder Polyisobutylen (B) ein Molekulargewicht nach dem Zahlenmittel von 200 bis 3.000 und eine Viskosität von 300 bis 50.000 cps hat.

30 5. Gestreckter Schichtfilm nach Anspruch 1, dadurch gekennzeichnet, daß besagter Polyglycerin-Fettsäureester (C) eine Verbindung ist, die durch Veresterung wenigstens einer Hydroxylgruppe eines Glycerinkondensationsprodukts mit einer höheren Fettsäure mit 8 bis 22 Kohlenstoffatomen erhalten wird.

35 6. Gestreckter Schichtfilm nach Anspruch 1, dadurch gekennzeichnet, daß besagte Komponente (D) ein gesättigter Polyester, hergestellt aus Terephthalsäure oder einem Derivat derselben und Ethylenglykol, ist.

40 7. Gestreckter Schichtfilm nach Anspruch 1, dadurch gekennzeichnet, daß er des weiteren eine Adhäsionsschicht zwischen besagter Zwischenschicht und besagter äußerer Schicht und zwischen besagter Zwischenschicht und besagter innerer Schicht umfaßt.

45 8. Gestreckter Schichtfilm nach Anspruch 7, dadurch gekennzeichnet, daß besagte Adhäsionsschicht modifizierte Polyolefine umfaßt.

9. Gestreckter Schichtfilm nach Anspruch 1, dadurch gekennzeichnet, daß besagter Film ein koextrudierter Schichtfilm ist.

45 10. Gestreckter Schichtfilm nach Anspruch 1, dadurch gekennzeichnet, daß besagter Film erhalten wird durch biaxiales Strecken eines nichtgestreckten Schichtfilms in der Maschinenrichtung und in der Querrichtung.

50 11. Gestreckter Schichtfilm nach Anspruch 1, dadurch gekennzeichnet, daß besagter Film erhalten wird durch einaxiales Strecken eines nichtgestreckten Schichtfilms in der Maschinenrichtung.

#### Revendications

55 1. Film stratifié et étiré comprenant au moins une couche extérieure, une couche intermédiaire et une couche intérieure,

ces couches extérieure et intérieure comprenant une composition à base de polypropylènes contenant 100 parties en poids de polypropylène (A), de 2 à 30 parties en poids de polybutène et/ou de polyisobutylène (B), et de 0,2 à 5 parties en poids d'un ester de type polyglycérol-acide gras (C), et

la couche intermédiaire comprenant des polyesters (D) contenant des motifs téraphthalate d'éthylène comme principal composant, ce film étant étiré uniaxiallement ou biaxiallement selon un taux d'étirage de 2 à 6 fois dans chaque direction ; et ce film ayant :

une résistance à l'arrachement par cisaillement de la couche extérieure et de la couche intérieure, non inférieure à 0,4 kg/cm<sup>2</sup>, et une résistance à l'arrachement à 180°, non supérieure à 4 g/50 mm, mesurée ainsi que cela est mentionné dans la description,

un allongement à la rupture par traction dans le sens machine non supérieur à 200 %, mesuré selon la norme JIS C 2318,

un rapport de l'allongement à la rupture par traction dans le sens machine à celui dans la direction transversale, non supérieur à 3,

une épaisseur non supérieure à 40 µm, et

une épaisseur de la couche intermédiaire de 10 à 90 % par rapport à l'épaisseur totale de film.

2. Film stratifié et étiré selon la revendication 1, les polypropylènes (A) comprenant un homopolymère de propylène et un copolymère de propylène.
3. Film stratifié et étiré selon la revendication 1, dans lequel les polypropylènes (A) ont un indice de fluidité à chaud de 0,5 à 20 g/10 mn, et une masse volumique de 0,89 à 0,91 g/cm<sup>3</sup>.
4. Film stratifié et étiré selon la revendication 1, dans lequel le polybutène et/ou le polyisobutylène (B), ont un poids moléculaire moyen en nombre de 200 à 3 000, et une viscosité de 300 à 50 000 cP.
5. Film stratifié et étiré selon la revendication 1, dans lequel l'ester de type polyglycérol-acide gras (C), est un composé obtenu par estérification d'au moins un groupe hydroxyle d'un produit de condensation du glycérol avec un acide gras supérieur comportant de 8 à 22 atomes de carbone.
6. Film stratifié et étiré selon la revendication 1, dans lequel le composant (D) est un polyester saturé produit à partir d'acide téraphthalique ou d'un dérivé de celui-ci, et d'éthylène glycol.
7. Film stratifié et étiré selon la revendication 1, comprenant en outre une couche adhésive entre la couche intermédiaire et la couche extérieure, et entre la couche intermédiaire et la couche intérieure.
8. Film stratifié et étiré selon la revendication 7, dans lequel la couche adhésive comprend des polyoléfines modifiées.
9. Film stratifié et étiré selon la revendication 1, qui est un film stratifié et coextrudé.
10. Film stratifié et étiré selon la revendication 1, obtenu par étirage biaxial d'un film stratifié et non étiré, dans le sens machine et dans la direction transversale.
11. Film stratifié et étiré selon la revendication 1, obtenu par étirage uniaxial d'un film stratifié et non étiré, dans le sens machine.

45

50

55

THIS PAGE BLANK (USPTO)